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INTERFERENCE PHENOMENA IN ATOMIC EMISSION NEAR AN INTERFACE: PURE CLASSICAL EFFECTS IN QUANTUM RADIATION

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ABSTRACT

The emission of fluorescence by an atom which is close to the surface of an optically-reflecting medium is examined. All linear media, like a dielectric, a metal, a thin film on a substrate, etc., are dealt with in a unified way. It appears that only the classical Fresnel coefficients for reflection of a plane wave enter the expression for the radiation field. The reflection by nonlinear media is also considered, and in particular optical phase conjugators which operate via degenerate four-wave mixing. The two strong laser beams which pump the nonlinear crystal are assumed to be in resonance with an atomic transition frequency. It is shown that the angular intensity distribution and the polarization of the fluorescence are determined by classical interference of waves, although the radiation is pure quantum mechanical. It is also shown that an atom in its ground state can fluoresce when it is near a phase conjugator, and that this phenomenon follows from the classical properties of this quantum radiation field.

Keywords: fluorescence, interface, phase conjugation, angular spectrum

I. INTRODUCTION

Emission of fluorescence radiation by an atom is a pure quantum mechanical process. An intriguing question is whether this underlying quantum mechanism in the production of the radiation is reflected in special properties of the emitted electromagnetic field, or, is the field after it is emitted first ordinary radiation, which is indistinguishable from a classical electromagnetic wave. The pioneering experiments by Kimble, Dagenais and Mandel[1-3] have shown that quantum radiation is indeed different, and that the mechanism of emission leaves its traces in the form of some peculiar statistical features in the sequence of photon detection from this radiation field. They found that fluorescence photons exhibit a so-called 'antibunching' behavior, which means that these photons arrive at a photomultiplier tube well-separated in time, e.g. two photons never arrive together. Photons in an electromagnetic field with a classical analogue, however, tend to stick together (they are 'bunched').

After acknowledging that fluorescence radiation is essentially quantum mechanical in nature, we can reverse the question. Which properties of the emitted field originate from quantum mechanics, and which properties have a simple explanation in classical terms. In this paper we consider the emission of fluorescence by an atom (dipole) which is positioned in the vicinity of a surface. The general approach to this problem[4-5] is to quantize explicitly the radiation field in a plane-wave mode decomposition, both above the surface and in the medium, and then calculate radiative lifetimes of excited states and fluorescent spectra. Recently we have shown,[6] however, that the structure of the spontaneous-decay operator for this situation is entirely determined by the rotational symmetry of the system under rotation about the normal direction to the surface, rather than by the nature of the quantized field for a specific medium. Here we present a derivation of the form of the radiation field, emitted by a dipole near a surface, but without reference to detailed properties of the medium, and without an explicit quantization of the field. Therefore, the results hold for both a quantum and a classical dipole, and in this way we can keep track of the classical features of this quantum radiation field. It also enables us to treat the various possible media with a unified theory.

An atom with a dipole-moment operator $\underline{\mu}$ is located at a distance h above a surface. We take the surface as the xy-plane and the empty space above it as $z > 0$. Then the position vector of the atom is $\underline{h}$ =

$h\mathbf{e}_z$. In the region $z < 0$ we have material like a dielectric layer, a metal (mirror), or a thin film on a substrate. The medium is infinite in extend in the x and y directions, compared to h. All these configurations have in common that the material is linear in its optical response, and therefore an incident plane wave will always be reflected in the specular direction. We shall consider separately the case where the medium is an optical phase conjugator based on four-wave mixing in a transparent medium.[7] The only difference is that a reflected wave in the region $z > 0$ now propagates in the direction opposite to the incident wave. We shall show that this seemingly minor difference in the optical response of the medium has dramatic consequences for the emission of fluorescence. Figure 1 illustrates the two situations.

II. GENERAL FORM OF THE ELECTRIC FIELD

The emitted fluorescence can be detected by a photomultiplier tube (either operating as a photon counter or as an intensity meter), which is sensitive to the electric component $\mathbf{E}(\mathbf{r},t)$ of the field. Since this detector is located in the region $z > 0$ above the surface, we shall restrict our attention to the solution for the field $\mathbf{E}(\mathbf{r},t)$ in this part of space only. In general, we can write for $z > 0$

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_v(\mathbf{r},t) + \mathbf{E}_p(\mathbf{r},t) + \mathbf{E}_h(\mathbf{r},t) \quad , \qquad (2.1)$$

where v, p and h stand for vacuum, particular and homogeneous, respectively. The vacuum field $\mathbf{E}_v$ is by definition the solution for the field $\mathbf{E}$ for the situation that the atom would not be there. In a quantum approach, this is the always-present vacuum radiation field, but in a classical treatment this term is absent. This $\mathbf{E}_v$ is responsible for the spontaneous decay of an excited atomic state, which leads to the emission of the fluorescence. Notice that this $\mathbf{E}_v$ must be different from the $\mathbf{E}_v$ for completely empty space, since the presence of the surface and the medium put limitations on the possible plane-wave solutions, due to the boundary conditions for the field at the surface. Also, $\mathbf{E}_v$ contains possible other freely-propagating components, like a laser field which can be applied to drive a certain atomic transition. Continuous excitation and spontaneous decay then gives rise to the emission of resonance fluorescence. The second term, $\mathbf{E}_p$, in Eq. (1.1) is the field of a dipole in $\mathbf{r} = \mathbf{h}$, but in empty space (e.g., no medium present). By adding the third term $\mathbf{E}_h$, which must be a solution of the homogeneous Maxwell equations (no source terms), we can let the total field $\underset{\sim}{E}$ satisfy the boundary conditions at $z = 0$. Obviously, $\underset{\sim}{E}_h$ is the dipole radiation which is reflected by the surface, whereas $\underset{\sim}{E}_p$ serves as an incident field.

The field $\underset{\sim}{E}(\underset{\sim}{r},t)$, together with the magnetic field, must obey Maxwell's equations, both in a classical and a quantum approach. In the quantum theory, $\underset{\sim}{E}(\underset{\sim}{r},t)$ is an operator field, and the Heisenberg equations for the field operator are identical in form to the classical Maxwell equations.[8] Therefore, we can solve both problems simultaneously. The source term is of course the oscillating dipole $\underset{\sim}{\mu}(t)$, but we shall keep the time dependence of $\underset{\sim}{\mu}(t)$ unspecified. In this fashion, we can treat the spontaneous decay of an excited atom and a laser-driven atom (where the t-dependence of $\underset{\sim}{\mu}(t)$ is a forced oscillation with the laser frequency) on an equal footing. Notice that the t-dependence of $\underset{\sim}{\mu}(t)$ represents the Heisenberg representation of the Schrödinger operator $\underset{\sim}{\mu}$.

Maxwell's equations are most easily solved in the Fourier domain. We define

$$\hat{\underset{\sim}{E}}(\underset{\sim}{r},\omega) = \int_{-\infty}^{\infty} dt \; e^{i\omega t} \, \underset{\sim}{E}(\underset{\sim}{r},t) \quad , \tag{2.2}$$

with ω real, and similar definitions hold for other time-dependent quantities (most notably $\hat{\underset{\sim}{\mu}}(\omega)$). The field must be Hermitian

$$\underset{\sim}{E}(\underset{\sim}{r},t)^{\dagger} = \underset{\sim}{E}(\underset{\sim}{r},t) \quad , \tag{2.3}$$

which reads in the Fourier domain as

$$\underset{\sim}{E}(\underset{\sim}{r},\omega)^{\dagger} = \underset{\sim}{E}(\underset{\sim}{r},-\omega) \quad . \tag{2.4}$$

A convenient concept is the positive frequency part of the field, defined as

$$\underset{\sim}{E}^{(+)}(\underset{\sim}{r},t) = \frac{1}{2\pi} \int_{0}^{\infty} d\omega \; e^{-i\omega t} \, \hat{\underset{\sim}{E}}(\underset{\sim}{r},\omega) \quad . \tag{2.5}$$

Then the field itself is given by

$$\underset{\sim}{E}(\underset{\sim}{r},t) = \underset{\sim}{E}^{(+)}(\underset{\sim}{r},t) + \underset{\sim}{E}^{(-)}(\underset{\sim}{r},t) \quad , \tag{2.6}$$

with

$$\underline{E}^{(-)}(\underline{r},t) = (\underline{E}^{(+)}(\underline{r},t))^{\dagger} \tag{2.7}$$

as the negative frequency part. The advantage of the introduction of $\underline{E}^{(+)}$ follows from Eq. (2.5). As soon as we know $\hat{\underline{E}}(\underline{r},\omega)$ for positive ω only, then we know the entire field $\underline{E}(\underline{r},t)$, as reflected by Eqs. (2.6) and (2.7). Similar relations hold for $\underline{\mu}(t)$, $\hat{\underline{\mu}}(\omega)$ and $\underline{\mu}^{(+)}(t)$, since the dipole moment operator is also Hermitian.

III. ANGULAR SPECTRUM OF DIPOLE RADIATION

The component $\hat{\underline{E}}_p$ of the radiation is the field of a dipole $\hat{\underline{\mu}}(\omega)$ at $\underline{r} = \underline{h}$, and in its most concise form it is given by

$$\hat{\underline{E}}_p(\underline{r},\omega) = \frac{1}{4\pi\epsilon_o}(k^2\hat{\underline{\mu}}(\omega) + (\hat{\underline{\mu}}(\omega)\cdot\nabla)\nabla)G(\underline{r}-\underline{h},k) \tag{3.1}$$

for $\omega > 0$, and with $k = \omega/c$. With the Green's function for outgoing waves

$$G(\underline{r}-\underline{h},k) = \frac{e^{ik|\underline{r}-\underline{h}|}}{|\underline{r}-\underline{h}|} \quad , \tag{3.2}$$

it is easy to verify that Eq. (3.1) is identical to the more familiar forms.[9]

In order to find the homogeneous contribution $\hat{\underline{E}}_h$ to the radiation field, which is the reflected field, we have to construct $\hat{\underline{E}}_p + \hat{\underline{E}}_h$ and something similar for the field inside the medium, and for the magnetic field. Then the boundary conditions which follow from Maxwell's equations determine the field everywhere in space. However, this is an extremely complicated procedure, due to the complexity of the dipole field. Besides that, this procedure depends on the nature of the medium. A thin layer on a metal substrate and a half-finite dielectric will yield very different results. With this in mind, we have developed a general technique to solve this problem, which is also directly applicable to nonlinear interactions like reflections at a phase conjugator.

As a first step, we use Weyl's representation of the Green's function, instead of the standard form (3.2). It can be shown[10] that Eq. (3.2) is equivalent to

$$G(\underset{\sim}{r}-\underset{\sim}{h},k) = \frac{i}{2\pi}\int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\, \frac{1}{\gamma}\, e^{i\underset{\sim}{K}_{\parallel}\cdot\underset{\sim}{r}+i\gamma|z-h|} \quad , \tag{3.3}$$

where the vector $\underset{\sim}{K}_{\parallel}$ is defined as

$$\underset{\sim}{K}_{\parallel} = \alpha\underset{\sim}{e}_x + \beta\underset{\sim}{e}_y \quad , \tag{3.4}$$

in terms of the integration variables α and β. The subscript $\parallel$ is a reminder that this vector is parallel to the surface, and for later purposes we define two other vectors in terms of $\underset{\sim}{K}_{\parallel}$ as

$$\underset{\sim}{K}_{\pm} = \underset{\sim}{K}_{\parallel} \pm \gamma\underset{\sim}{e}_z \quad . \tag{3.5}$$

The parameter γ is given by

$$\gamma = \sqrt{k^2-\alpha^2-\beta^2+i\delta} \quad , \quad \delta \downarrow 0 \quad , \tag{3.6}$$

where the notation $\delta \downarrow 0$ implies that the imaginary part of γ is positive.

Now we can substitute expression (3.3) for the Green's function into Eq. (3.1) and carry out the differentiations. After some rearrangement of terms, we obtain the representation for the particular solution as

$$\begin{aligned}
\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = &- \frac{1}{\epsilon_o}\hat{\mu}_z(\omega)\underset{\sim}{e}_z\delta(\underset{\sim}{r}-\underset{\sim}{h}) \\
&+ \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\, \frac{i}{8\pi^2\epsilon_o\gamma}\, e^{i\underset{\sim}{K}_{\parallel}\cdot\underset{\sim}{r}+i\gamma|z-h|} \\
&\times [(-\gamma(\underset{\sim}{K}_{\parallel}\cdot\hat{\underset{\sim}{\mu}}(\omega))\mathrm{sgn}(z-h) + K_{\parallel}^2\hat{\mu}_z(\omega))\underset{\sim}{e}_z \\
&+ k^2\hat{\underset{\sim}{\mu}}_{\parallel}(\omega) - (\underset{\sim}{K}_{\parallel}\cdot\hat{\underset{\sim}{\mu}}(\omega) + \gamma\hat{\mu}_z(\omega)\mathrm{sgn}(z-h))\underset{\sim}{K}_{\parallel}] \quad ,
\end{aligned} \tag{3.7}$$

where $\hat{\underset{\sim}{\mu}}_{\parallel}$ and $\hat{\mu}_z\underset{\sim}{e}_z$ are the parallel and perpendicular components of $\hat{\underset{\sim}{\mu}}$ with respect to the surface, respectively. Expression (3.7) is sometimes called the angular spectrum of plane waves. It simply means that for every combination of α and β the integrand of Eq. (3.7) is a plane wave, and it is easy to check that these waves are transverse. For $\gamma > 0$ we have travelling waves, but for γ imaginary the waves are evanescent, e.g., decaying in the +z and -z directions. Also notice that the waves are different for $z > h$ and $0 < z < h$, due to the appearance of $\mathrm{sgn}(z-h)$ and $|z-h|$. This guarantees that all plane-wave components emanate from the position $\underset{\sim}{r} = \underset{\sim}{h}$ of the atom.

IV. DECOMPOSITION INTO s AND p WAVES

Although the angular representation of dipole radiation, Eq. (3.7), still looks very complicated, it is now obvious where the simplification enters. The double integral is a summation over transverse plane-wave components, which satisfy Maxwell's equations individually. In order to find the reflected dipole field, we only have to know how a classical plane wave is reflected. By superimposing the various components in the same way as in Eq. (3.7), we then obtain immediately $\hat{\underset{\sim}{E}}_h(\underset{\sim}{r},\omega)$.

The amplitude ratio of the reflected and the incident wave is usually expressed in terms of Fresnel coefficients, which are different for s (surface) and p (plane) polarized waves. Therefore, we first have to decompose the angular spectrum into s and p waves. We see that all plane waves in Eq. (3.7) have either wave vector $\underset{\sim}{K}_+$ or $\underset{\sim}{K}_-$, depending on the sign of z-h. For the s and p unit polarization vectors for a given wave vector, we adopt the following phase convention:

$$\underset{\sim}{e}_{\underset{\sim}{K}_\pm s} = \frac{1}{K_\parallel}\,\underset{\sim}{K}_\parallel \times \underset{\sim}{e}_z \quad , \tag{4.1}$$

$$\underset{\sim}{e}_{\underset{\sim}{K}_\pm p} = \frac{1}{kK_\parallel}\,(\pm\gamma\underset{\sim}{K}_\parallel - K_\parallel^2\underset{\sim}{e}_z) \quad . \tag{4.2}$$

For $z > h$ the wave vector of any plane wave is given by $\underset{\sim}{K}_+$, as follows from Eq. (3.7), and therefore we have to decompose the factor in square brackets into an $\underset{\sim}{e}_{\underset{\sim}{K}_+ s}$ and an $\underset{\sim}{e}_{\underset{\sim}{K}_+ p}$ component. The remarkably simple result is

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\; e^{i\underset{\sim}{K}_+\cdot\underset{\sim}{r}}\, \frac{ik^2 e^{-i\gamma h}}{8\pi^2\epsilon_o\gamma} \times \sum_\sigma (\underset{\sim}{e}_{\underset{\sim}{K}_+\sigma}\cdot\hat{\underset{\sim}{\mu}}(\omega))\underset{\sim}{e}_{\underset{\sim}{K}_+\sigma} \quad , \qquad z > h \quad , \tag{4.3}$$

where the summation over σ runs over σ = s and p. Every plane wave $\exp(i\underset{\sim}{K}_+\cdot\underset{\sim}{r})$ is either travelling or decaying in the +z direction, and they correspond to the (d) photons from Fig. 1.

The waves which travel into the direction of the surface are found to be

$$\hat{\underset{\sim}{E}}_p(\underset{\sim}{r},\omega) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta \; e^{i\underset{\sim}{K}_-\cdot\underset{\sim}{r}} \frac{ik^2 e^{i\gamma h}}{8\pi^2\epsilon_o\gamma} \sum_\sigma (\underset{\sim}{e}_{\underset{\sim}{K}_-\sigma}\cdot\hat{\underset{\sim}{\mu}}(\omega))\underset{\sim}{e}_{\underset{\sim}{K}_-\sigma} \quad ,$$

$$0 < z < h \quad , \qquad (4.4)$$

which are of the $\exp(i\underset{\sim}{K}_-\cdot\underset{\sim}{r})$ type. These plane waves serve as the incident waves on the surface, for which we have to find the (classical) reflection coefficients.

V. REFLECTED FIELD

In order to obtain the angular representation of the homogeneous solution $\hat{\underset{\sim}{E}}_h$, we recognize that Eq. (4.4) gives the plane-wave expansion of the incident field, so we only have to find the reflected waves on a per-wave basis. We can proceed along these lines in a general way, provided that we distinguish between two cases.

V.a. *Linear Medium*

As illustrated in Fig. 1, for a linear medium the reflected wave is always in the specular direction, no matter the kind of medium. We found in the previous section that the incident waves have wave vector $\underset{\sim}{K}_-$ and polarization vector $\underset{\sim}{e}_{\underset{\sim}{K}_-\sigma}$. But then the reflected wave simply has wave vector $\underset{\sim}{K}_+$, and for the phase convention of the polarization we can again take $\underset{\sim}{e}_{\underset{\sim}{K}_+\sigma}$ as polarization vectors. We indicate the Fresnel coefficient for reflection of a $\underset{\sim}{K}_-,\sigma$ wave by $R_{\underset{\sim}{K}_-\sigma}$. Then the total reflected field is immediately found to be

$$\hat{\underset{\sim}{E}}_h(\underset{\sim}{r},\omega) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta \; e^{i\underset{\sim}{K}_+\cdot\underset{\sim}{r}} \frac{ik^2 e^{i\gamma h}}{8\pi^2\epsilon_o\gamma} \sum_\sigma R_{\underset{\sim}{K}_-\sigma}(\underset{\sim}{e}_{\underset{\sim}{K}_-\sigma}\cdot\hat{\underset{\sim}{\mu}}(\omega))\underset{\sim}{e}_{\underset{\sim}{K}_+\sigma} \quad ,$$

$$z > 0. \qquad (5.1)$$

Although expansion (4.4) for the incident waves only holds for $0 < z < h$, the solution (5.1) pertains, of course, to the entire region $z > 0$.

V.b. *Nonlinear Transparent Medium*

If the dielectric constant of the material in $z < 0$ almost equals unity (transparent medium), then the specularly-reflected wave is absent. The only way a reflected wave can be generated is via the third-order susceptibility (for isotropic media). But $\chi^{(3)}$ is about 20 orders of magnitude smaller than $\chi^{(1)}$, and hence this contribution to reflected radiation is also negligible. However, there is a way of enhancing the third-order interaction considerably, without introducing a first-order (linear) interaction. If we shine two strong counterpropagating laser beams through the material, and in a direction parallel to the surface, then these two fields couple to the incident field via $\chi^{(3)}$. Effectively, this enhances the coupling parameter for the nonlinear interaction by a factor which is proportional to the intensity of the two pump beams (supposed to be equal). With contemporary high-power lasers it is easy to achieve an interaction strength which produces reflected radiation with an intensity comparable to the intensity of the incident wave, or even higher.[11]

In the described setup, the device (medium plus two pump lasers) operates as a phase conjugator.[12] Without going into the details of the mechanism of optical phase conjugation, it is easy to show[7] that in general the wave vector of the reflected light is opposite to the wave vector of the incident wave, as shown in Fig. 1. This implies that the reflected wave retraces the path of the incident wave, and so it travels back to the atom which emitted the wave. In this sense, optical phase conjugation is identical to time reversal. A slightly more careful analysis[13] shows that in fact the negative frequency component of the reflected field is proportional to the positive frequency part of the incident wave, where the ratio of amplitudes is again a simple Fresnel reflection coefficient, and both waves have the same wave vector.[14] For classical radiation this subtlety makes no difference, but for quantum radiation it has dramatic consequences, as we shall show later in the paper. Then, with Eq. (4.4) we can construct the homogeneous solution

$$\underline{E}_h(\underline{r},-\omega) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\, e^{i\underline{K}\cdot\underline{r}}\, \frac{ik^2 e^{i\gamma h}}{8\pi^2\epsilon_o\gamma} \sum_{\sigma} P_{\underline{K}\sigma}(\underline{e}_{\underline{K}\sigma}\cdot\underline{\hat{\mu}}(\omega))\underline{e}_{\underline{K}\sigma} \quad , \qquad (5.2)$$

where $P_{\underline{K}\sigma}$ indicates the Fresnel coefficient for reflection at the PC.

Equation (5.2) holds for $\omega > 0$, so this gives $\underset{\sim}{\hat{E}}_h$ for negative frequencies. The positive frequency components can then be found by Hermitian conjugation, according to Eq. (2.4). This yields[15]

$$\underset{\sim}{\hat{E}}_h(\underset{\sim}{r},\omega) = \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\, e^{-i\underset{\sim}{K}_- \cdot \underset{\sim}{r}} \frac{ik^2 e^{-i\gamma h}}{8\pi^2 \epsilon_o \gamma}$$

$$\times \sum_{\sigma} P^{*}_{\underset{\sim}{K}_-\sigma} (\underset{\sim}{e}_{\underset{\sim}{K}_-\sigma} \cdot \underset{\sim}{\hat{\mu}}(-\omega)) \underset{\sim}{e}_{\underset{\sim}{K}_-\sigma} , \tag{5.3}$$

where we used $\underset{\sim}{\hat{\mu}}(\omega)^{\dagger} = \underset{\sim}{\hat{\mu}}(-\omega)$.

VI. FLUORESCENCE NEAR A LINEAR MEDIUM

The results from the previous sections give the Fourier transform of the radiation field in the entire region $z > 0$, but the angular-spectrum representation is cumbersome. Fortunately, it is not necessary to know the field everywhere in space, since the detectors which measure the radiation are macroscopic devices at a large distance from the atom. Consequently, we only have to evaluate the field more explicitly for distances r between atom and detector, which are much larger than the wavelength of the radiation. Both the waves which travel directly to the detector, Eq. (4.3), and waves which are first reflected by the surface, Eq. (5.1), are of the $\exp(i\underset{\sim}{K}_+ \cdot \underset{\sim}{r})$ type. With the method of stationary phase[16] the general form of an asymptotic expansion of an angular spectrum is found to be

$$\frac{i}{2\pi} \int_{-\infty}^{\infty} d\alpha \int_{-\infty}^{\infty} d\beta\, \frac{1}{\gamma}\, e^{i\underset{\sim}{K}_+ \cdot \underset{\sim}{r}}\, h(\alpha,\beta) \sim \frac{e^{ikr}}{r}\, h(\alpha_o,\beta_o) \tag{6.1}$$

for r large and any function $h(\alpha,\beta)$. Here, α_o and β_o are

$$\alpha_o = k \sin\theta\cos\phi \quad , \quad \beta_o = k \sin\theta\sin\phi \quad , \tag{6.2}$$

with (θ,ϕ) the spherical coordinates of the position of the detector with respect to the position of the atom (or origin).

Now we can make an asymptotic expansion of the fields $\underset{\sim}{\hat{E}}_p$ and $\underset{\sim}{\hat{E}}_h$ from Eqs. (4.3) and (5.1), respectively. For the particular solution we find

$$\underline{\hat{E}}_p(\underline{r},\omega) = \frac{\omega^2}{4\pi\epsilon_0 c^2 r} e^{ik(r-h\cos\theta)}([\underline{e}_\vartheta\cdot\underline{\hat{\mu}}(\omega)]\underline{e}_\vartheta + [\underline{e}_\phi\cdot\underline{\hat{\mu}}(\omega)]\underline{e}_\phi) \quad , \quad (6.3)$$

in terms of the spherical unit vectors $\underline{e}_\vartheta$ and $\underline{e}_\phi$ for the direction (ϑ,ϕ). For the reflected field we obtain

$$\underline{\hat{E}}_h(\underline{r},\omega) = \frac{\omega^2}{4\pi\epsilon_o c^2 r} e^{ik(r+h\cos\theta)} (R_p(\theta)[\underline{e}_\vartheta\cdot\underline{\hat{\mu}}'(\omega)]\underline{e}_\vartheta$$

$$- R_s(\theta)[\underline{e}_\phi\cdot\underline{\hat{\mu}}'(\omega)]\underline{e}_\phi) \quad , \qquad (6.4)$$

where

$$\underline{\hat{\mu}}'(\omega) = \underline{\hat{\mu}}(\omega)_\perp - \underline{\hat{\mu}}(\omega)_\parallel \qquad (6.5)$$

in terms of the perpendicular and parallel components of the dipole operator $\underline{\hat{\mu}}(\omega)$ with respect to the surface. This $\underline{\hat{\mu}}(\omega)$ has the significance of the mirror image of $\underline{\hat{\mu}}$, although the surface is not necessarily a mirror. The difference between a perfect mirror and an arbitrary surface is taken into account by the Fresnel coefficients $R_\sigma(\theta)$. These appearing Fresnel coefficients are the reflection coefficients for an incident wave which would be scattered in the observation direction (θ,ϕ). It follows from symmetry that $R_\sigma(\theta)$ is independent of the angle ϕ. Comparison of Eqs. (6.3) and (6.4) shows that the effective distance between the source and the detector is $r-h\cos\theta$ and $r+h\cos\theta$, repectively. The difference is $2h\cos\theta$, which is exactly the difference in path length that a directly-emitted and a reflected photon has to travel, in order to arrive at the detector at an angle θ. As illustrated in Fig. 2., it seems that the reflected photon comes from a mirror dipole, a distance h below the surface.

Now we can add $\underline{\hat{E}}_p$ and $\underline{\hat{E}}_h$ in order to find the total radiation field in the far zone (neglecting the vacuum field, since no photons can be detected in this field). Subsequently we take the Fourier inverse, under the assumption that the radiation has a narrow frequency width around a certain frequency ω_o. For an atom, this is the transition frequency between two levels. We then obtain for the positive frequency part

$$\underline{E}^{(+)}(\underline{r},t) = \frac{\omega_o^2}{4\pi\epsilon_o c^2 r} \sum_\sigma [\underline{e}_\sigma \cdot \underline{m}(t-r/c,\theta,\phi)]\underline{e}_\sigma \quad , \qquad (6.6)$$

where the summation runs over $\sigma = \theta,\phi$. The vector operator $\underline{m}$ is an effective dipole moment operator for the combination atom plus surface, and it is explicitly

$$\underline{m}(t,\theta,\phi) = e^{-i\omega_o\tau}\,\underline{\mu}^{(+)}(t) + e^{i\omega_o\tau}\{R_p(\theta)[\underline{e}_\theta\cdot\underline{\mu}^{(+)}(t)']\underline{e}_\theta$$
$$- R_s(\theta)[\underline{e}_\phi\cdot\underline{\mu}^{(+)}(t)']\underline{e}_\phi\} \quad , \qquad (6.7)$$

where

$$\tau = h\cos\theta/c \qquad (6.8)$$

equals half the difference in retardation time between a directly-emitted and a reflected photon. Equations (6.6) and (6.7) contain all information about the intensity distribution and the polarization of the fluorescence radiation.

VII. FLUORESCENCE NEAR A PC

The radiation field in the far zone for emission near a PC can be found along the same lines as in the previous section. We now find

$$\underline{E}^{(+)}(\underline{r},t) = \frac{\omega_o^2}{4\pi\epsilon_o c^2 r} \sum_\sigma [\underline{e}_\sigma \cdot \underline{M}(t-r/c,\theta,\phi)]\underline{e}_\sigma \quad , \qquad (7.1)$$

where the effective dipole operator for the entire system is now given by

$$\underline{M}(t,\theta,\phi) = e^{-i\omega_o\tau}[\underline{\mu}^{(+)}(t) - e^{-2i\omega_o t}\{P_p^*(\theta)[\underline{e}_\theta\cdot\underline{\mu}^{(-)}(t)]\underline{e}_\theta$$
$$+ P_s^*(\theta)[\underline{e}_\phi\cdot\underline{\mu}^{(-)}(t)]\underline{e}_\phi\}] \quad . \qquad (7.2)$$

The first term in square brackets is again the directly-emitted fluorescence. We notice that the second term now depends on the negative-frequency component of the dipole operator, rather than its positive-frequency part. The factor $\exp(-2i\omega_o t)$ makes the total term again a positive-frequency field. Another important difference with Eq. (6.7) is that both terms in Eq. (7.1) have the same retardation factor $\exp(-i\omega_o\tau)$. Therefore, it seems that the phase-conjugated photons are effectively emitted from the site of the atom, and not from a mirror atom below the surface. We shall see in Sec. IX that this has far-reaching consequences for emission by a quantum dipole.

VIII. INTENSITY DISTRIBUTION

A photodetector in the radiation zone (r large) can measure the fluorescence radiation. The photon counting rate from any electric field $\underset{\sim}{E}$, in number of photons per unit time per unit solid angle Ω, is given by[17]

$$\frac{\partial^2 W}{\partial t \partial \Omega} = 2\epsilon_o c r^2 \langle \underset{\sim}{E}^{(-)}(\underset{\sim}{r},t)\cdot\underset{\sim}{E}^{(+)}(\underset{\sim}{r},t)\rangle \quad , \tag{8.1}$$

where the angle brackets indicate either a quantum expectation value or an average over a possible stochasticity of a classical radiation field. For the problem at hand we have expressions (6.6) and (7.1) for $\underset{\sim}{E}^{(+)}$, which are identical in form. For radiative emission near a linear medium we then find with Eqs. (6.6) and (8.1)

$$\frac{\partial^2 W}{\partial t \partial \Omega} = \frac{\omega_o^4}{8\pi^2 \epsilon_o c^3} \sum_\sigma \langle [\underset{\sim}{e}_\sigma \cdot \underset{\sim}{m}(t-r/c,\theta,\phi)^\dagger][\underset{\sim}{e}_\sigma \cdot \underset{\sim}{m}(t-r/c,\theta,\phi)]\rangle \quad , \tag{8.2}$$

and for a nonlinear medium we simply replace $\underset{\sim}{m}$ by $\underset{\sim}{M}$.

Now we can substitute the explicit forms (6.7) and (7.2) for $\underset{\sim}{m}$ and $\underset{\sim}{M}$, respectively, into Eq. (8.2), and work out the various products. At this stage it is imperative to realize that $\underset{\sim}{m}$ and $\underset{\sim}{M}$ represent quantum operators in general, and that an operator product is not necessarily commutative. We obtain for the linear case

$$\frac{\partial^2 W}{\partial t \partial \Omega} = \frac{\omega_o^4}{8\pi^2 \epsilon_o c^3} \sum_\sigma (1 + |R_\sigma(\theta)|^2)$$

$$\times \langle [\underset{\sim}{e}_\sigma \cdot \underset{\sim}{\mu}^{(-)}(t-r/c)][\underset{\sim}{e}_\sigma \cdot \underset{\sim}{\mu}^{(+)}(t-r/c)]\rangle \quad , \tag{8.3}$$

and for reflection at a PC

$$\frac{\partial^2 W}{\partial t \partial \Omega} = \frac{\omega_o^4}{8\pi^2 \epsilon_o c^3} \sum_\sigma (<[\underline{e}_\sigma \cdot \underline{\mu}^{(-)}(t-r/c)][\underline{e}_\sigma \cdot \underline{\mu}^{(+)}(t-r/c)]>$$

$$+ |P_\sigma(\theta)|^2 <[\underline{e}_\sigma \cdot \underline{\mu}^{(+)}(t-r/c)][\underline{e}_\sigma \cdot \underline{\mu}^{(-)}(t-r/c)]>) \quad . \qquad (8.4)$$

Apart from possibly different values for the reflection coefficients R_σ and P_σ, the only difference between Eqs. (8.3) and (8.4) is that in the last term of Eq. (8.4) the order of $\underline{\mu}^{(+)}$ and $\underline{\mu}^{(-)}$ is reversed, as compared to Eq. (8.3). For a classical dipole, both $\underline{\mu}^{(+)}$ and $\underline{\mu}^{(-)}$ are vectors in ordinary space, and products of their components commute. We conclude that the angular intensity distribution of the radiation for a classical dipole near a PC is indistinguishable from the radiation pattern of that same dipole near a linear medium. In particular, for a perfect mirror we have $|R_\sigma(\theta)| = 1$, and for a perfect phase-conjugating mirror we have $|P_\sigma(\theta)| = 1$, which makes Eqs. (8.3) and (8.4) identical. For a quantum dipole (an atom), however, expressions (8.3) and (8.4) can render quite different results, as we shall show in the next section.

The total emission rate of photons into the half space $z > 0$ above the surface is given by

$$\frac{dW}{dt} = \int_{z>0} d\Omega \, \frac{\partial^2 W}{\partial t \partial \Omega} \quad . \qquad (8.5)$$

It is possible to evaluate this intensity of emission explicitly in terms of angular integrals over the reflection coefficients $|R_\sigma(\theta)|^2$ and $|P_\sigma(\theta)|^2$, weighted with a certain factor, but for the purpose of illustration we make a simplifying assumption. Suppose that $|R_\sigma(\theta)|^2$ and $|P_\sigma(\theta)|^2$ are independent of the polarization σ, and independent of the angle of incidence θ, as for a perfect mirror and a perfect PC. Then the integration over the solid angle Ω in Eq. (8.5) can be performed easily for the two cases where expressions (8.3) and (8.4) appear in the integrand. The result for a linear medium is

$$\frac{dW}{dt} = \frac{\omega_o^4}{6\pi\epsilon_o c^3} (1 + |R|^2) <\underline{\mu}^{(-)}(t-r/c) \cdot \underline{\mu}^{(+)}(t-r/c)> \quad , \qquad (8.6)$$

and for a PC we obtain

$$\frac{dW}{dt} = \frac{\omega_o^4}{6\pi\epsilon_o c^3} \{<\underset{\sim}{\mu}^{(-)}(t-r/c)\cdot\underset{\sim}{\mu}^{(+)}(t-r/c)>$$

$$+ |P|^2<\underset{\sim}{\mu}^{(+)}(t-r/c)\cdot\underset{\sim}{\mu}^{(-)}(t-r/c)>\} \quad . \tag{8.7}$$

Again, the only difference in form between Eqs. (8.6) and (8.7) is that $\underset{\sim}{\mu}^{(+)}\cdot\underset{\sim}{\mu}^{(-)}$ is not necessarily equal to $\underset{\sim}{\mu}^{(-)}\cdot\underset{\sim}{\mu}^{(+)}$.

XI. TWO-LEVEL ATOM

As an important example we consider a non-degenerate two level atom with excited state $|e>$ and ground state $|g>$. The energy separation between the two levels is $\hbar\omega_o$, and the matrix element of the transition dipole moment between the levels is $\underset{\sim}{\mu}_{ge} = <g|\underset{\sim}{\mu}|e>$. Then the positive-frequency part $\underset{\sim}{\mu}^{(+)}(t)$ of the dipole operator is the Heisenberg representation of the Schrödinger operator

$$\underset{\sim}{\mu}^{(+)} = |g>\underset{\sim}{\mu}_{ge}<e| \quad , \tag{9.1}$$

which is proportional to the atomic lowering operator $|g><e|$. Similarly, we have $\underset{\sim}{\mu}^{(-)} = (\underset{\sim}{\mu}^{(+)})^\dagger = |e>\underset{\sim}{\mu}^*_{ge}<g|$. If we want to evaluate the right-hand sides of Eqs. (8.6) and (8.7), then we have to transform the expressions first to the Schrödinger representation, and subsequently calculate the expectation value for a given atomic wave function. It is not necessary to know explicitly this wave function. The general result is

$$\frac{dW}{dt} = \frac{1}{2}A\hbar\omega_o(1 + |R|^2)n_e(t-r/c) \tag{9.2}$$

for a linear medium, and

$$\frac{dW}{dt} = \frac{1}{2}A\hbar\omega_o[n_e(t-r/c) + |P|^2n_g(t-r/c)] \tag{9.3}$$

for a PC. Here, n_e and n_g are the populations of the excited and ground state, respectively, at the retarded time $t - r/c$. Their values depend on the preparation of the atom in a certain state. If we pass an atomic beam with ground-state atoms over the surface of the medium, then at a certain moment a strong laser pulse can partially excite the atoms, and with a proper choice of pulse shape and intensity the atoms can be prepared in virtually any desired state. The only restriction is

$$n_e(t) + n_g(t) = 1 \tag{9.4}$$

at all times.

Now let us look at the interpretation of Eqs. (9.2) and (9.3). First, recall that the emission rate for this atom in empty space is given by $dW/dt = A\hbar\omega_o n_e(t-r/c)$, with

$$A = \frac{\omega_o^3}{3\pi\epsilon_o\hbar c^3} |\underline{\mu}_{ge}|^2 \tag{9.5}$$

as the Einstein coefficient for spontaneous decay from level $|e\rangle$ to level $|g\rangle$. For an atom at $\underline{r} = \underline{h}$, half the amount of photons will be emitted in the direction $z > 0$, and the other half in the direction $z < 0$. Inspection of Eqs. (9.2) and (9.3) shows that the first term in both cases is exactly $\frac{1}{2}A\hbar\omega_o n_e(t-r/c)$, which is half the emission rate in free space. We conclude that these first terms coresond to photons which are emitted directly into the direction $z > 0$, as if there were no surface at all. The other half of the number of photons are emitted into the direction $z < 0$, and they travel towards the surface. For a linear medium they have a probability of $|R|^2$ to be reflected, just like a classical wave, and hence the number of photons which will travel towards the detector in $z > 0$ is exactly $|R|^2$ times the number of photons that was emitted into the direction $z < 0$. This gives the second term in Eq. (9.2). Figure 2 illustrates the situation.

Also for a PC, the number of photons per unit time that is emitted into the direction $z < 0$ equals $\frac{1}{2}A\hbar\omega_o n_e(t-r/c)$, but Eq. (9.3) shows that there is not a second term, like in Eq. (9.2), which accounts for the fraction of reflected photons. There are at least two possible explanations for this peculiar fact. First, the dipole radiation, which is emitted towards the PC, is a spherical diverging wave. For a classical wave of this kind, the reflected and conjugated wave would be a converging spherical wave which travels towards the atom, and is focused exactly on the atom. After the emission, the atom was left in its ground state, so when the conjugated photon comes back, it can be absorbed again by the atom (stimulated excitation). In that process the atom would return to its original excited state. In this picture, photons bounce back and forth between the atom and the PC, and effectively the atom remains in its excited state. A second explanation would be that the photons which hit the PC are simply absorbed, and do not come back at all. In that case, the atom would be left in its ground state after the emission, because of energy considerations. It is impossible to decide which interpretation is correct. This would require at least a quantum description of the state of the atom as a function of time, so that we can follow the time evolution of $n_e(t)$ during the emission.

Another kind of strange behavior is reflected by the appearance of the second term in Eq. (9.3). This term is proportional to the population n_g of the ground state. If the atom would be entirely in its ground state ($n_e = 0$, $n_g = 1$), it emits fluorescent photons at a rate $\frac{1}{4}A\hbar\omega_o|P|^2$ into the region $z > 0$. A possible interpretation of this phenomenon is the following. The presence of the dipole polarizes the PC, which subsequently emits spontaneously a fluorescent photon, focused on the atom. The atom absorbs the photon, goes to its excited state, and decays again. During this regular decay it emits a photon directly into the direction of the detector. The net effect for the atom is that it remains in its ground state. The atom merely acts as a medium which extracts photons from the PC. Notice that this process has no classical analogue, since there is a 'reflected' field, but without an incident field. Figure 3 visualizes some possible interpretations of the behavior of an atom near a PC. We emphasize that these explanations are tentative, and that more complicated schemes are possible.

X. CONCLUSIONS

We have studied the emission of radiation by a dipole near a surface, where we allowed the reflecting medium to be either linear or nonlinear. Since the Heisenberg equations for the electromagnetic field operators are identical in form as the classical Maxwell equations, we were able to treat both the emission by a classical and by a quantum dipole within the same framework. A consequence of this analogy is that a major part of the features of a quantum radiation field is entirely classical. We have shown that also the reflected quantum field can be expressed in terms of the classical Fresnel coefficients for reflection, and that the details of the structure of the medium are irrelevant for the form of the radiation field. All that has to be known are these Fresnel coefficients.

Classical interference between the quantum waves determines the intensity distribution and polarization of the radiation in the far zone. Even the total emission rate dW/dt can be be found without an explicit quantization of the field, and the modifications due to the presence of the surface can be accounted for by the Fresnel coefficients. Even more obscure phenomena, like those predicted by Eq. (9.3), have their origin in simple classical interference between the various waves. We only needed the fact that $\underline{\mu}$ is an atomic operator, but no reference to the quantum aspects of the radiation was required for the derivation of this result.

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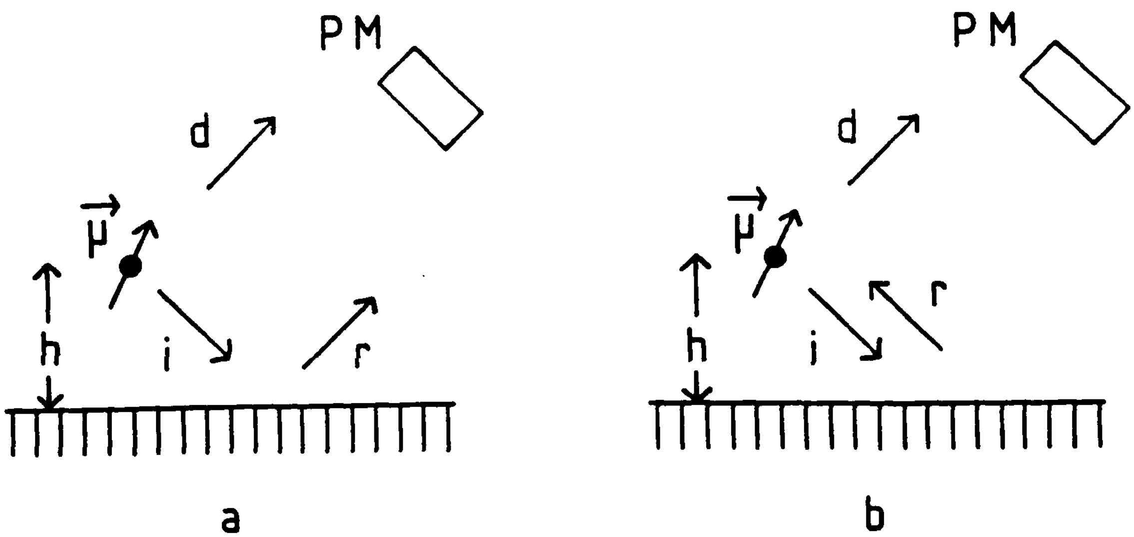

Fig. 1. Reflection of an incident (i) plane-wave component of the fluorescence, emitted by the dipole $\underline{\mu}$, at a linear (a) and a nonlinear transparent (b) medium. The only distinction is the propagation direction of the reflected (r) wave. The waves or photons labeled (d) propagate directly towards the photomultiplier (PM).

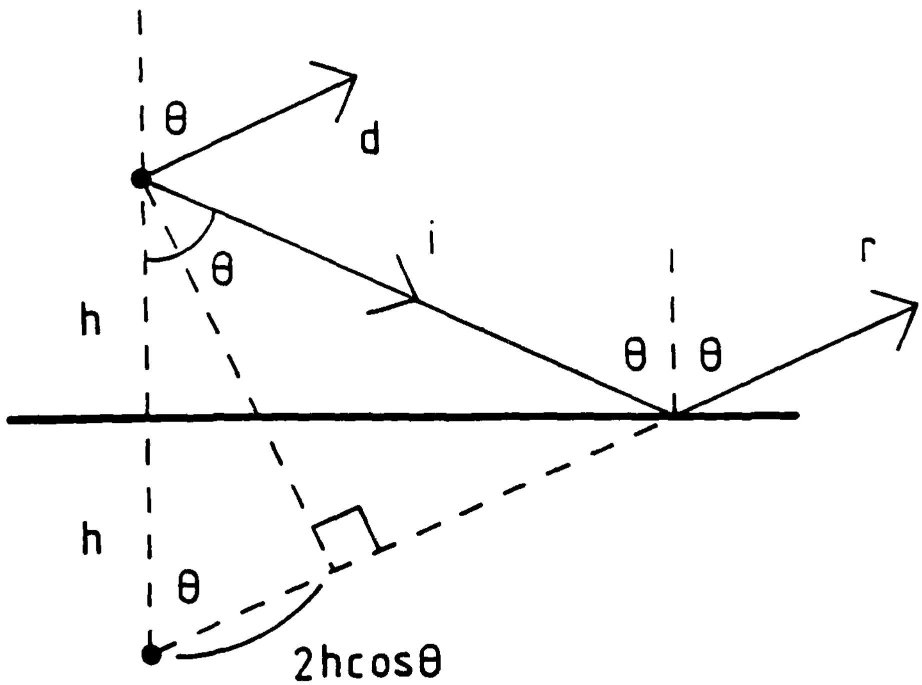

Fig. 2. A detector is positioned at an angle θ with the z-axis. Photons which arrive at a certain time can have travelled two different paths. They can either go directly (d) from the atom to the detector, or they can be scattered by the (linear) surface first (i plus r path). The difference in path length is 2hcosθ, and the difference in travel time is 2τ. It seems that the r-photons are emitted by a mirror dipole, a distance h below the surface. Also notice that the angle of incidence equals the angle of reflection, just as for classical fields.

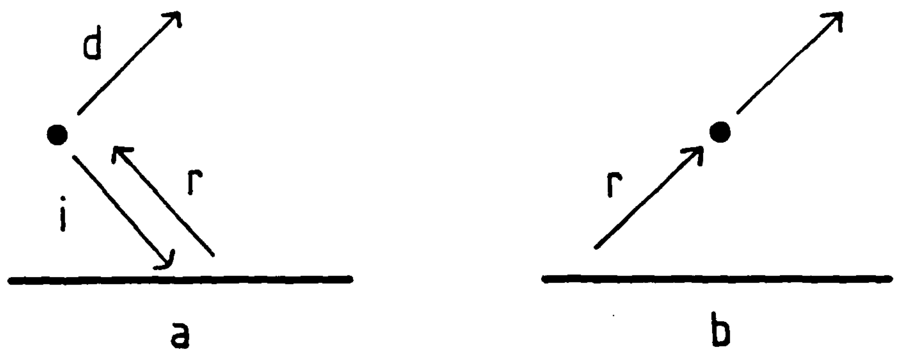

Fig. 3. Different mechanisms for the emission of fluorescence near a PC. In (a), the atom is in its excited state, and only the directly-emitted (d) photons end up in the detector. The i-photons interact with the PC, and are either completely absorbed or reflected back. If the latter would be the case, then the atom absorbs the r-photon again. No r-photon can pass the atom in case (a). In (b), the atom is in its ground state. The PC spontaneously emits a photon, which is absorbed by the atom, and subsequently emitted into the direction of the detector.

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